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(54) Rubber composition.

(57) There is disclosed a rubber composition comprising:

(A) 100 parts by weight of a fluororubber;

(B) 2 to 200 parts by weight of a fluorosilicone gum that contains siloxane units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, in which Me denotes a methyl radical;(C) 0.01 to 40 parts by weight of a fluorosilicone oil comprising siloxane units of the formula $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ and siloxane units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, wherein a is an integer having a value of at least 4, n is 0, 1, or 2 and Me denotes a methyl radical;

(D) 1 to 100 parts by weight of a silica micropowder; and

(E) a curing agent in a quantity sufficient to cure the composition, wherein the fluorosilicone oil comprising component (C) imparts an excellent compatibility between fluororubber (A) and fluorosilicone gum (B).

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The invention relates to a rubber composition comprising a fluororubber, a fluorosilicone gum, a fluorosilicone oil, silica and a curing agent.

Fluororubbers are characterized by excellent heat, oil, and weather resistance and by excellent mechanical strength. As a result, they are used in applications where such properties are critical. However, fluororubbers have poor, low-temperature flexibility. Fluororubbers have been mixed with fluorosilicone rubbers, which have excellent low-temperature properties, in an attempt to correct this deficiency in JP-A(s) 56-50948 and 63-213545. However, the poor compatibility between fluororubbers and fluorosilicone rubbers has made it difficult to obtain uniform mixtures by simply mixing the two. The resulting mixtures also have very unsatisfactory processabilities. JP-A 2-196838 has proposed the addition of epoxy-functional organosiloxane to the fluororubber and fluorosilicone rubber mixture to solve these problems. The resulting rubber, however, has poor mold-release characteristics, and this particular deficiency imposes restrictions on its range of application.

We have found unexpectedly that the addition of a special fluorosilicone oil as a compatibilizer for a mixture of fluororubber and fluorosilicone rubber results in enhanced compatibility and also provides cured rubber moldings that have good mold-release along with excellent physical properties.

This invention introduces a rubber composition that cures to give rubber moldings with good mold-release and excellent physical properties, said composition comprising

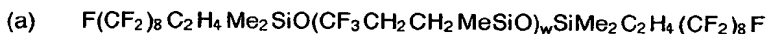
- (A) 100 weight parts of a fluororubber;
- (B) 2 to 200 weight parts of a fluorosilicone gum that contains the $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ siloxane unit, wherein Me denotes a methyl group;
- (C) 0.01 to 40 weight parts of a fluorosilicone oil that contains the $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ siloxane unit, in which a is an integer having a value of at least 4 and n is 0, 1, or 2, and the $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ siloxane unit;
- (D) 1 to 100 weight parts of a silica micropowder; and
- (E) a curing agent, in a quantity sufficient to cure the composition.

The fluororubber, component (A), used by the present invention comprises fluorine-containing rubbery elastomers, with organoperoxide-vulcanizable fluororubbers being preferred. Fluororubber is exemplified by vinylidene fluoride homopolymers, vinylidene fluoride-tetrafluoroethylene copolymers, vinylidene fluoride-hexafluoropropylene copolymers, vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymers, tetrafluoroethylene-hexafluoropropylene copolymers, and tetrafluoroethylene-perfluoroalkyl vinyl ether copolymers. Among these, fluororubber that contains a crosslinkable functional group (crosslink site monomer) is preferred.

The fluorosilicone gum, component (B), is an organopolysiloxane containing the $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ siloxane unit (Me = methyl), and this siloxane unit preferably makes up at least 70 mole% of component (B). Siloxane units that may be present in component (B) in addition to the $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ unit are by methylvinylsiloxane, allylmethylsiloxane, hexenylmethylsiloxane, dimethylsiloxane, methylphenylsiloxane, and methylethylsiloxane. The methylvinylsiloxane unit is preferred. In addition to the preceding siloxane units, the siloxane unit $\text{RSiO}_{3/2}$ (R = monovalent hydrocarbon group) may make up part of the siloxane units other than $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$. The monovalent hydrocarbon group R is exemplified by alkyl groups, such as methyl and ethyl; alkenyl groups, such as vinyl and allyl; aryl groups, such as phenyl; and the 3,3,3-trifluoropropyl group. The terminal group of component (B) is exemplified by dimethylvinylsiloxy, trimethylsiloxy, methylvinylphenylsiloxy, silanol, 3,3,3-trifluoropropyldimethylsiloxy, and 3,3,3-trifluoropropylmethylvinylsiloxy. Component (B) typically has a degree of polymerization (DP) of 1,000 to 20,000. Component (B) is added at 2 to 200 weight parts per 100 weight parts component (A), and preferably at 5 to 150 weight parts per 100 weight parts component (A).

The fluorosilicone oil component (C) is the component that characterizes the composition of the invention. Component (C) functions as a compatibilizer during the mixing of components (A) and (B) to improve compatibility between them. Component (C) consists of organopolysiloxane that contains the $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ siloxane unit and the $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ siloxane unit. The subscript a is an integer with a value of at least 4 (for example, 4, 6, and 8), and a need not have only a single value. The subscript n is 0, 1, or 2, and Me denotes the methyl group. The $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ unit to $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ unit molar ratio is preferably from 1:9 to 9:1. Component (C) may be a random or block copolymer, with block copolymers being preferred. Alkyl groups are methyl, ethyl, and propyl, and alkenyl groups such as vinyl, allyl, and so forth, are Examples of the organic groups that may be bonded in any siloxane units present in addition to the two units specified above. Methyl is preferred. The molecular structure of component (C) is exemplified by straight-chain, branched, and cyclic structures. Component (C) preferably has a viscosity at 25 °C in the range of 10 to 10,000 mPa.s (centipoise).

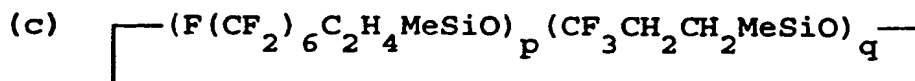
Fluorosilicone oil (C) is exemplified by fluorosilicone oils with the following general formulas (a), (b), and (c).



(w is an integer with a value of 1 to 300 and Me = methyl)



(x and y are each integers with values of at least 1,
(x + y) = 2 to 1,000, and Me = methyl)



(p and q are each integers with values of at least 1,
(p + q) = 3 to 30, and Me = methyl)

These fluorosilicone oils may be used individually or in combinations of two or more.

Fluorosilicone oil with general formula (a) may be synthesized by the following methods:

- (i) hydrolysis of heptadecafluorodecyldimethylchlorosilane by itself, followed by polymerization with cyclic 3,3,3-trifluoropropylmethylsiloxane in the presence of polymerization catalyst; or
- (ii) cohydrolysis of heptadecafluorodecyldimethylchlorosilane with 3,3,3-trifluoropropylmethyldichlorosilane, followed by polymerization with cyclic 3,3,3-trifluoropropylmethylsiloxane in the presence of polymerization catalyst.

Fluorosilicone oil with general formula (b) may be synthesized by the following methods:

- (i) cohydrolysis of nonafluorohexylmethyldichlorosilane with 3,3,3-trifluoropropylmethyldichlorosilane, followed by re-equilibration copolymerization in the presence of hexamethyldisiloxane using a known basic catalyst;
- (ii) dehydrative polycondensation between the alpha,omega-dihydroxypoly(nonafluorohexylmethylsiloxane) and alpha,omega-dihydroxypoly(3,3,3-trifluoropropylmethylsiloxane) produced by the hydrolysis, respectively, of nonafluorohexylmethyldichlorosilane by itself and 3,3,3-trifluoropropylmethyldichlorosilane by itself;
- (iii) chain elongation between the alpha,omega-dihydroxypoly(nonafluorohexylmethylsiloxane) and alpha,omega-dihydroxypoly(3,3,3-trifluoropropylmethylsiloxane) produced above, using a crosslinker such as difunctional organosilane; and
- (iv) living polymerization, using sodium silanolate or lithium silanolate, of a mixture of nonafluorohexylmethylcyclotrisiloxane and 3,3,3-trifluoropropylmethylcyclotrisiloxane.

These reactions may be heated as desirable and may be run in organic solvent.

Fluorosilicone oil of general formula (c) may be synthesized by the following methods:

- (i) cohydrolysis of tridecafluorooctylmethyldichlorosilane with 3,3,3-trifluoropropylmethyldichlorosilane;
- (ii) subjecting the alpha,omega-dihydroxypoly(tridecafluorooctylmethylsiloxane-co-3,3,3-trifluoropropylmethylsiloxane) yielded by the preceding method to re-equilibration in the presence of a known basic catalyst;
- (iii) dehydrative polycondensation between the alpha,omega-dihydroxypoly(tridecafluorooctylmethylsiloxane) and alpha,omega-dihydroxypoly(3,3,3-trifluoropropylmethylsiloxane) produced by the hydrolysis, respectively, of tridecafluorooctylmethyldichlorosilane by itself and 3,3,3-trifluoropropylmethyldichlorosilane by itself; and
- (iv) chain elongation between the alpha,omega-dihydroxypoly(tridecafluorooctylmethylsiloxane) and alpha,omega-dihydroxypoly(3,3,3-trifluoropropylmethylsiloxane) produced above, using a crosslinker such as difunctional organosilane.

The preceding reactions may be run in organic solvent.

Component (C) is added at 0.01 to 40 weight parts, preferably at 0.1 to 20 weight parts, per 100 weight parts of component (A). The cured product yielded by the composition has reduced physical properties at additions in excess of 40 weight parts. At the other extreme, the compatibility between components (A) and (B) declines at component (C) additions below 0.01 weight part.

The silica micropowder component (D) is a reinforcing filler whose purpose is to increase the mechanical strength of the composition. This component should have a specific surface area of at least 50 m²/g, and preferably has a specific surface area of 100 to 400 m²/g. Usable as component (D) are those silica micropowders already known for use as fillers for fluorosilicone rubbers. For example, the fumed silicas and calcined silicas obtained by dry processes and the precipitated silicas obtained by wet processes are suitable. Also usable as component (D) are the hydrophobic silicas obtained by hydrophobizing the surface of the aforementioned silicas by treatment with an organosilicon compound such as dimethyldichlorosilane, hexamethyldisilazane, diphenylsilanediol, and 1,3-bis(3,3,3-trifluoropropyl)-tetramethyldisilazane. Component (D) is added at a level of 1 to 100 weight parts, and preferably 5 to 70 weight parts, per 100 weight parts of component (A).

The curing agent, component (E), functions to cure the composition of our invention, and any curing agent may be used that is able to cure components (A) and (B). This component encompasses those curing agents already known in the art as curing agents for fluorosilicone rubber compositions. Organoperoxides are preferred including benzoyl peroxide, 2,4-dichlorobenzoyl peroxide, di-tert-butyl peroxide, tert-butylmonochlorobenzoyl peroxide, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane, and the like. Component (E) should be added in a quantity sufficient to induce the cure of the invention composition, and in general is added at 0.1 to 20 weight parts per 100 weight parts of component (A).

In addition to components (A) to (E), various additives known for use in fluororubbers and fluorosilicone rubbers may also be used insofar as the object of this invention is not impaired. Examples of such additives are wetting agents, extending fillers, heat stabilizers, vulcanization aides, softeners, plasticizers, antioxidants, and vulcanization accelerators. Wetting agents improve the affinity between the fluorosilicone gum (B) and silica micropowder (D), and are exemplified by silanol-endblocked diorganopolysiloxanes having a low DP. The silicon-bonded organic groups in this diorganopolysiloxane are exemplified by alkyl groups such as methyl, ethyl, and propyl; alkenyl groups such as vinyl and allyl; aryl groups such as phenyl; and 3,3,3-trifluoropropyl. Silanol-endblocked 3,3,3-trifluoropropylmethylsiloxanes are specifically preferred. This siloxane preferably has a DP in the range of 1 to 100, preferably of 2 to 30. Extending fillers are exemplified by quartz powder, diatomaceous earth, calcium carbonate, aluminosilicates, mica, talc, and aluminum oxide. Heat stabilizers are exemplified by iron oxide, cerium oxide, cerium hydroxide, titanium oxide, aluminum oxide, zinc oxide, manganese carbonate, and carbon black. Triallyl isocyanurate is an example of a vulcanization aide.

The rubber composition of this invention is prepared by mixing components (A) to (E) to homogeneity, or by blending any additives into these components and then mixing to homogeneity. For example, components (A) to (D) may be mixed in a kneader mixer for 3 to 20 minutes with heating and the curing agent (E) can then be mixed in on a two-roll mill. Suitable mixers are exemplified by kneader mixers, ram cover-equipped kneader mixers, Banbury™ mixers, continuous mixers, two-roll mills, and three-roll mills.

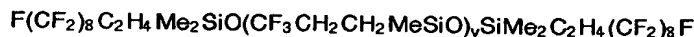
The presence of the specific fluorosilicone oil (C) in the rubber composition of our invention induces compatibility between the fluororubber (A) and fluorosilicone gum (B) to the level of homogeneity. This prevents any decline in physical properties due to compromised compatibility and thereby yields cured rubber moldings with excellent physical properties. These moldings also exhibit excellent mold-releasability. Finally, since our claimed composition exhibits the highly desirable properties of both the fluororubber (A) and the fluorosilicone gum (B), its cured rubber moldings exhibit excellent resistance to cold, heat, solvent, and oil.

Examples

The invention is explained in greater detail through working examples. In the examples and comparative example, "part" denotes "weight parts", the viscosity is the value at 25 °C, Me stands for methyl, Vi stands for vinyl, and cP is an abbreviation for centipoise.

Example 1

The following were charged to a kneader mixer and mixed to homogeneity: 70 parts of a fluororubber in the form of a vinylidene fluoride-hexafluoropropylene copolymer, 22.2 parts of dimethylvinylsiloxane-endblocked 3,3,3-trifluoropropylmethylsiloxane-methylvinylsiloxane copolymer gum (average DP = 7,000) composed of 99.5 mole% siloxane unit with the formula CF₃CH₂CH₂MeSiO and 0.5 mole% siloxane unit with the formula CH₃ViSiO, 1 part fluorosilicone oil (viscosity = 50 mPa.s/cP) with the following formula



(v is a number that yields a viscosity of 50 mPa.s/cP),

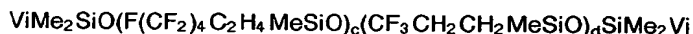
7.5 parts of a surface-hydrophobicized fumed silica (specific surface area = 200 m²/g), and 0.3 part of cerium oxide as a heat stabilizer. 100 parts of this mixture was then mixed to homogeneity on a two-roll mill with 0.5 part of 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane to yield a curable rubber composition. The rubber composition was compression molded for 10 minutes at 170 °C/30 kg/cm² to yield a 2 mm-thick rubber sheet. Curing was brought to completion by heating the rubber sheet in an oven for an additional 4 hours at 200 °C. The physical properties of the resulting rubber sheet (hardness, tensile strength, elongation, and tear strength) were measured in accordance with JIS K 6301, and the measurement results are reported in Table 1 below. The rubber sheet exhibited very good mold-release properties. In addition, the rubber sheet gave a smooth fracture surface, which was indicative of a thorough dispersion of each component in the rubber composition.

Comparative Example 1

A rubber composition was prepared as in Example 1, but in this case omitting the 1 part fluorosilicone oil that was used in Example 1. The resulting rubber composition was compression molded as in Example 1 to give a 2 mm-thick rubber sheet, whose cure was again brought to completion by heating in an oven at 200 °C for an additional 4 hours. The physical properties of the resulting rubber sheet are reported in Table 1 below.

Example 2

The following were charged to a kneader mixer and mixed to homogeneity: 50 parts of a fluororubber in the form of a vinylidene fluoride-hexafluoropropylene copolymer, 37 parts of a dimethylvinylsiloxane-endblocked 3,3,3-trifluoropropylmethyl-siloxane-methylvinylsiloxane copolymer gum (average DP = 7,000) composed of 99.5 mole% siloxane unit with the formula CF₃CH₂CH₂MeSiO and 0.5 mole% siloxane unit with the formula CH₃ViSiO, 1.2 parts of a fluorosilicone oil comprising block copolymer (viscosity = 1,000 mPa.s/cP) with the following formula



(c:d = 1:1),

12.5 parts of a surface-hydrophobicized fumed silica (specific surface area = 200 m²/g), and 0.5 part of cerium oxide as heat stabilizer. 100 parts of this mixture was then mixed to homogeneity on a two-roll mill with 0.5 part of 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane to yield a curable rubber composition. The rubber composition was compression molded for 10 minutes at 170 °C and 3.0 mPa pressure (30 kg/cm²) to yield a 2 mm-thick rubber sheet. Curing was brought to completion by heating the rubber sheet in an oven for an additional 4 hours at 200 °C. The physical properties of the resulting rubber sheet are reported in Table 1. The rubber sheet exhibited very good mold-release properties. In addition, the rubber sheet gave a smooth fracture surface, which was indicative of a thorough dispersion of each component in the cured rubber composition.

Example 3

The following were charged to a kneader mixer and mixed to homogeneity: 100 parts of a dimethylvinylsiloxane-endblocked 3,3,3-trifluoropropylmethylsiloxane-methylvinylsiloxane copolymer gum (average DP = 7,000) composed of 99.0 mole% siloxane unit with the formula CF₃CH₂CH₂MeSiO and 1.0 mole% siloxane unit with the formula CH₃ViSiO, 8 parts of a silanol-endblocked 3,3,3-trifluoropropylmethylsiloxane with average DP = 5, and 35 parts of a fumed silica with a specific surface area of 300 m²/g. To the resulting mixture was added 50 parts of a fluororubber in the form of a vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymer and 5 parts of a fluorosilicone oil (number average molecular weight = 30,000) with the following formula



(e:f = 1:4),

followed by mixing for 1 hour. 100 parts of this mixture were then mixed to homogeneity on a two-roll mill

with 0.5 part of 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane to yield a curable rubber composition. The rubber composition was compression molded for 10 minutes at 170 °C and 3.0 mPa pressure (30 kg/cm²) to yield a 2 mm-thick rubber sheet. Curing was brought to completion by heating the rubber sheet in an oven for an additional 4 hours at 200 °C. The physical properties of the resulting rubber sheet are also reported in Table 1. The rubber sheet exhibited very good mold-release properties. In addition, the rubber sheet gave a smooth fracture surface, which was indicative of a thorough dispersion of each component in the cured rubber composition.

Table 1

	Example 1	Example 2	Example 3	Comparative Example 1
hardness (JIS A)	73	74	64	72
tensile strength (kg/cm ²)	125	113	116	89
[mPa]	[12.5]	[11.3]	[11.6]	[8.9]
elongation (%)	280	340	350	230
tear strength (kg/cm)	23	21	21	19
[kN/M]	[22.5]	[20.6]	[20.6]	[18.6]

Claims

1. A rubber composition comprising:

- (A) 100 parts by weight of a fluororubber;
- (B) 2 to 200 parts by weight of a fluorosilicone gum that contains siloxane units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, in which Me denotes a methyl radical;
- (C) 0.01 to 40 parts by weight of a fluorosilicone oil comprising siloxane units of the formula $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ and siloxane units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, wherein a is an integer having a value of at least 4, n is 0, 1, or 2 and Me denotes a methyl radical;
- (D) 1 to 100 parts by weight of a silica micropowder; and
- (E) 0.1 to 20 parts by weight of a curing agent sufficient to cure the composition.

2. The composition according to claim 1, wherein said fluororubber (A) is selected from the group consisting of vinylidene fluoride homopolymers, vinylidene fluoride-tetrafluoroethylene copolymers, vinylidene fluoride-hexafluoropropylene copolymers, vinylidene fluoride-tetrafluoroethylene-hexafluoropropylene copolymers, tetrafluoroethylene-hexafluoropropylene copolymers, and tetrafluoroethylene-perfluoroalkyl vinyl ether copolymers.

3. The composition according to claim 2, wherein at least 70 mole% of the siloxane units of said fluorosilicone gum (B) are units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, in which Me denotes a methyl radical.

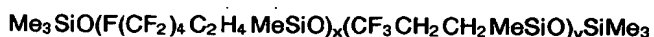
4. The composition according to claim 3, wherein the molar ratio of $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ units to $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$ units is 1:9 to 9:1, wherein Me denotes a methyl radical.

5. The composition according to claims 1 or 4, wherein said fluorosilicone oil (C) has the formula



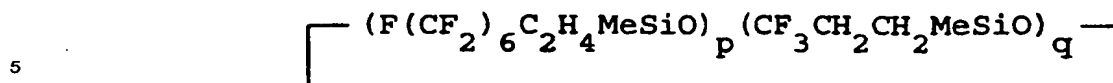
in which w is an integer having a value of 1 to 300 and Me denotes a methyl radical.

6. The composition according to claims 1 or 4, wherein said fluorosilicone oil (C) has the formula



in which x and y are each integers with values of at least 1, $(x + y) = 2$ to 1,000 and Me denotes a methyl radical.

7. The composition according to claims 1 or 4, wherein said fluorosilicone oil (C) has the formula



in which p and q are each integers with values of at least 1, $(p + q) = 3$ to 30 and Me denotes a methyl radical.

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8. The composition according to claim 4, wherein said silica (D) has a specific surface area of 100 to 400 m²/gram and is present at a level of 5 to 70 parts by weight for each 100 parts by weight of said fluororubber (A).

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9. The composition according to claim 8, wherein said fluorosilicone gum (B) is present at a level of 5 to 150 parts by weight and said fluorosilicone oil (C) is present at a level of 0.1 to 20 parts by weight for each 100 parts by weight of said fluororubber (A).

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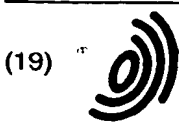
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(54) **Rubber composition**

(57) There is disclosed a rubber composition comprising:

- (A) 100 parts by weight of a fluororubber;
- (B) 2 to 200 parts by weight of a fluorosilicone gum that contains siloxane units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, in which Me denotes a methyl radical;
- (C) 0.01 to 40 parts by weight of a fluorosilicone oil comprising siloxane units of the formula $\text{F}(\text{CF}_2)_a\text{CH}_2\text{CH}_2\text{Me}_n\text{SiO}_{(3-n)/2}$ and siloxane units of the formula $\text{CF}_3\text{CH}_2\text{CH}_2\text{MeSiO}$, wherein a is an integer having a value of at least 4, n is 0, 1, or 2 and Me denotes a methyl radical;
- (D) 1 to 100 parts by weight of a silica micropowder; and
- (E) a curing agent in a quantity sufficient to cure the composition, wherein the fluorosilicone oil comprising component (C) imparts an excellent compatibility between fluororubber (A) and fluorosilicone gum (B).

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European Patent
Office

EUROPEAN SEARCH REPORT

Application Number
EP 94 11 8777

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
D,A	EP-A-0 380 104 (DOW CORNING TORAY SILICONE) 1 August 1990 * page 2, line 22 - page 3, line 29 *	1-9	C08K3/36 C08L27/12 C08L83/08 //(C08L27/12, 83:08,83:08), (C08L83/08, 27:12,83:08)
A	GB-A-1 284 081 (RAYCHEM) 2 August 1972 * examples 6,7 *	1-9	
A	US-A-3 192 175 (J.R.RUSSELL) 29 June 1965 * column 1, line 29 - line 38 *	1-9	
A	WO-A-81 00573 (MINNESOTA MINING & MFG) 5 March 1981 * page 4, line 6 - line 12 *	1-9	
The present search report has been drawn up for all claims			TECHNICAL FIELDS SEARCHED (Int.Cl.6)
			C08K C08L
Place of search THE HAGUE		Date of completion of the search 19 July 1996	Examiner Schmidt, H
<p>CATEGORY OF CITED DOCUMENTS</p> <p>X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document</p> <p>T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons</p> <p>Δ : member of the same patent family, corresponding document</p>			

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